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COMMENTS

Comment on "Pressure-induced amorphization of ZrTiCuNiBe bulk glass-forming alloy" [Appl. Phys. Lett. 79, 1106 (2001)]

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In a recent letter, Wang *et al.*[1] reported the pressure-induced amorphization of a $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ bulk glass-forming alloy. The concept of high-density and low-density state of the metallic glass was proposed and authors claim that accounts for observations of the differences in properties for samples prepared from water-quenched (alloy A) and under pressure (alloy B). However, Fig. 1 in the letter[1] clearly demonstrates that alloy B consists of two components: amorphous and crystalline components. The volume fraction of the crystalline component estimated by the authors in alloy B is about 10%. The crystallization process of the $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ bulk glass, which is one of the most studied bulk metallic glasses, has been intensively investigated during cooling from melt or heating from ambient temperature.[2,3] Schroers *et al.*[2] reported that the crystalline phase formed during cooling from melt is enriched in Zr and depleted in Be. The composition of the amorphous component in alloy B should differ from the initial alloy $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$. We, therefore, feel a need to provide the reader with a second explanation, different microstructures of alloy A and B, to the experimental results obtained in the letter.[1]

(1) The densities of alloy A and B are reported in the letter[1] to be 6.108 g/cm^3 and 6.181 g/cm^3, respectively. Alloy A, prepared by water-quenched method with a composition of $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$, is fully amorphous while alloy B is composed of amorphous and crystalline components. It is known that density of a material usually depends on composition and structure. The crystalline phase in alloy B could be higher than 6.108 g/cm^3 due to high content of Zr and low Be content. Furthermore, *just due to composition difference*, the density of the amorphous component in alloy B could also differ from 6.108 g/cm^3. Thus, the discrepancy in density can be attributed to different microstructures for alloy A and B.

(2) An enhancement of glass transition, T_g, and crystallization temperature, T_x, of alloy B is reported in the letter[1] as compared to alloy A. It has been demonstrated in many metallic glass systems that T_g and T_x strongly depend on composition. As we mentioned, the composition of the amorphous component in alloy B differs from that of alloy A. Consequently, different values of T_g and T_x should be expected in both alloys. This was actually observed in ZrTiCuNiBe alloy.[4] Furthermore, elastic properties and hardness of a material are, in general, sensitive to its microstructure. Thus, the changes observed for alloy A and B in velocities of transverse and longitudinal waves, shear modulus, Debye temperature, and hardness are not surprising and they are most likely also linked with different microstructures as mentioned above in the samples.

(3) Figure 1 in the letter[1] shows that the sample prepared under 1 atm. is fully crystallized while a 90% volume fraction of the amorphous component was detected in the samples prepared under pressures. Authors thus claim an enhancement of the glass forming ability for the $Zr_{41}Ti_{14}Cu_{12.5}Ni_{10}Be_{22.5}$ alloy under pressure. However, based on a broad experimental study on Zr-based metallic glasses under pressure using multianvil apparatus, the sample assembly is usually not tight when no pressure was applied. Oxides after solidification could be detected. On the other hand, under high hydrostatic pressure, (the minimum pressure required depends on the assembly size and condition), no oxides were detected, indicating that the heterogeneous nucleation becomes less significant. Therefore, we believe that no existence of the amorphous component in the sample under 1 atm could just be due to the heterogeneous nucleation effect while the formation of the amorphous component in the samples under pressure is resulted from less heterogeneous nucleation.

In conclusion, the experimental results presented in the letter[1] can be explained by different microstructures in the water-quenched sample and samples prepared under pressure. No clear evidence of the existence of the high-density state for the metallic glass was provided in the letter[1] although the high-density and low-density states in metallic glasses might exist.

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